

REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188		
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1. REPORT DATE (DD-MM-YYYY) 30-03-2015		2. REPORT TYPE Final Report		3. DATES COVERED (From - To) 15-Nov-2012 - 14-Feb-2015	
4. TITLE AND SUBTITLE Final Report: Enhanced Light Emitters based on Metamaterials			5a. CONTRACT NUMBER W911NF-13-1-0001		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER 611102		
6. AUTHORS Vinod Menon			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAMES AND ADDRESSES Queens College/Cuny And Research Founda 65-30 Kissena Blvd. Flushing, NY 11367 -1575			8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211			10. SPONSOR/MONITOR'S ACRONYM(S) ARO		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) 62509-EL.11		
12. DISTRIBUTION AVAILABILITY STATEMENT Approved for Public Release; Distribution Unlimited					
13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not contrued as an official Department of the Army position, policy or decision, unless so designated by other documentation.					
14. ABSTRACT We report the development of light emitters based on hyperbolic metamaterials. During the 18 month program period in Queens College of CUNY (Nov 2012 – May 2014), we successfully demonstrated growth of ultrasmooth silver films using germanium wetting layer, use of a high refractive index contrast grating to out-couple light from active hyperbolic metamaterials. We also successfully demonstrated for the first time simultaneous enhancement in spontaneous emission ad light extraction from active metamaterial structures.					
15. SUBJECT TERMS metamaterials, LEDs, microcavity					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Vinod Menon
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU			19b. TELEPHONE NUMBER 212-650-7443

Report Title

Final Report: Enhanced Light Emitters based on Metamaterials

ABSTRACT

We report the development of light emitters based on hyperbolic metamaterials. During the 18 month program period in Queens College of CUNY (Nov 2012 – May 2014), we successfully demonstrated growth of ultrasmooth silver films using germanium wetting layer, use of a high refractive index contrast grating to out-couple light from active hyperbolic metamaterials. We also successfully demonstrated for the first time simultaneous enhancement in spontaneous emission and light extraction from active metamaterial structures.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
03/29/2015 10.00	Xiaoze Liu, Tal Galfsky, Zheng Sun, Fengnian Xia, Erh-chen Lin, Yi-Hsien Lee, Stéphane Kéna-Cohen, Vinod M. Menon. Strong light-matter coupling in two-dimensional atomic crystals, Nature Photonics, (12 2014): 0. doi: 10.1038/nphoton.2014.304
03/29/2015 9.00	T. Galfsky, H. N. S. Krishnamoorthy, W. Newman, E. E. Narimanov, Z. Jacob, V. M. Menon. Active hyperbolic metamaterials: enhanced spontaneous emission and light extraction, Optica, (01 2015): 0. doi: 10.1364/OPTICA.2.000062
TOTAL:	2

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

<u>Received</u>	<u>Paper</u>
TOTAL:	

Number of Papers published in non peer-reviewed journals:

(c) Presentations

0

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

03/29/2015 2.00 Vinod M. Menon, Harish Krishnamoorthy, Zubin Jacob, Evgenii Narimanov, Ilona Kretzschmar. Broadband QED using Hyperbolic Metamaterials, Conference on Coherence and Quantum Optics. 12-AUG-13, Rochester, New York. : ,

03/29/2015 8.00 T. Galfsky, H. Krishnamoorthy, V.M. Menon, W. Newman, Z. Jacob, E. Narimanov. Extracting Light from High-K Modes in a Hyperbolic Metamaterial, 2014 IEEE Photonics Society Summer Topical Meeting Series. 14-JUL-14, Montreal, QC, Canada. : ,

03/29/2015 6.00 Xiaoze Liu, Tal Galfsky, Fengnian Xia, Erh-chen Lin, Yi-Hsien Lee, Ashwin Ramasubramaniam, Stephane Kena-Cohen, Vinod M. Menon. Strong light-matter coupling in atomic monolayers, CLEO: QELS_Fundamental Science. 08-JUN-14, San Jose, California. : ,

03/29/2015 5.00 Harish Krishnamoorthy, Ward D. Newman, Evgenii Narimanov, Zubin Jacob, Vinod M. Menon, Tal Galfsky. Directional emission from quantum dots in a hyperbolic metamaterial, CLEO: QELS_Fundamental Science. 08-JUN-14, San Jose, California. : ,

03/29/2015 4.00 H. N. S. Krishnamoorthy, Z. Jacob, T. Galfsky, E. Narimanov, I. Kretzschmar, V. M. Menon. Optical topological transition in metamaterials: QED and related effects, 2013 IEEE Photonics Conference (IPC). 08-SEP-13, Bellevue, WA, USA. : ,

08/30/2013 1.00 Zubin Jacob, Tal Galfsky, Evgenii E. Narimanov, Ilona Kretzschmar, Vinod M. Menon, Harish Krishnamoorthy. Topological Transitions in Metamaterials: QED and Related Effects, CLEO: QELS_Fundamental Science. 10-JUN-13, CLEO: QELS_Fundamental Science (2013). : ,

TOTAL: 6

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Book

TOTAL:

Received Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	Discipline
Tal Galfsky	1.00	
FTE Equivalent:	1.00	
Total Number:	1	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Vinod Menon	0.10	
FTE Equivalent:	0.10	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: 0.00

Names of Personnel receiving masters degrees

<u>NAME</u>
Total Number:

Names of personnel receiving PhDs

<u>NAME</u>

Total Number:

Names of other research staff

<u>NAME</u>

<u>PERCENT SUPPORTED</u>

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

The main scientific progress made during the program include:

- Realization of ultrasmooth sub-wavelength thick silver films for hyperbolic metamaterials
- Using high index contrast germanium gratings for light extraction
- Simultaneous enhancement in spontaneous emission rate and light extraction from active hyperbolic metamaterials.

Further details about the scientific accomplishments can be found in the attached document.

Technology Transfer

Report Type:	Final Report
Proposal Number:	62509EL
Agreement Number:	W911NF1310001
Proposal Title:	Enhanced Light Emitters based on Metamaterials
Report Period Begin Date:	11/15/2012
Report Period End Date:	05/31/2014

ABSTRACT

We report the development of light emitters based on hyperbolic metamaterials. During the 18 month program period in Queens College of CUNY (Nov 2012 – May 2014), we successfully demonstrated growth of ultrasmooth silver films using germanium wetting layer, use of a high refractive index contrast grating to out-couple light from active hyperbolic metamaterials. We also successfully demonstrated for the first time simultaneous enhancement in spontaneous emission and light extraction from active metamaterial structures.

OBJECTIVE

To utilize hyperbolic metamaterials (HMM) to enhance the performance of LEDs and realize sub-wavelength lasers. Specific goals include:

- Develop ultrafast and bright LEDs using hyperbolic metamaterials.
- Demonstrate enhancement of light emission from inherently low quantum efficiency emitters such as silicon nanocrystals.
- Develop sub-wavelength size lasers using emitters embedded in hyperbolic metamaterials.

1. INTRODUCTION

Under the proposed research program we are developing active HMMs comprising of one-dimensional metal-dielectric structures with colloidal quantum dots. Schematic drawing of the structure is shown in **Fig. 1**. The colloidal QDs are deposited via spin coating and capped with poly methyl methacrylate. The metal and dielectric layers are deposited via sputtering/electron beam evaporation. The gratings are defined via focused ion beam etching.

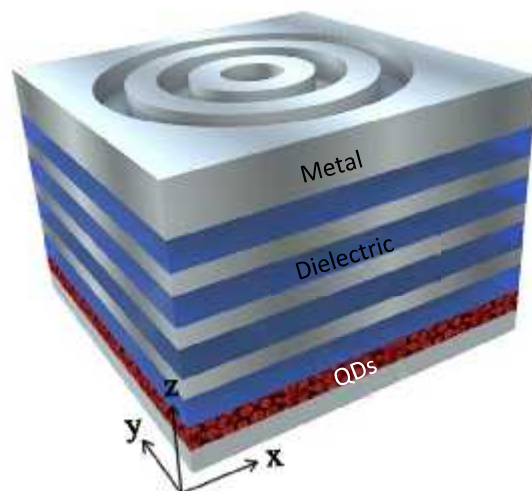


Fig. 1 Schematic of the metal dielectric metamaterial structure that results in the hyperbolic dispersion

In any medium, the optical iso-frequency curve $\tilde{S}(\vec{k}) = \text{const}$ can be engineered by tailoring the dielectric tensor $\tilde{V}(\vec{r})$. Metal dielectric composites can make the permittivity anisotropic and can considerably distort the topology of the iso-frequency curve. We consider the case of metal-dielectric composite metamaterials such as the one shown in **Fig. 1** which have a uniaxial form of the dielectric tensor $\tilde{\epsilon} = \text{diag}(\epsilon_{xx}, \epsilon_{xx}, \epsilon_z)$. The iso-frequency curve for the extraordinary (TM-polarized) waves propagating in such strongly anisotropic metamaterial is given by:

$$\frac{k_x^2 + k_y^2}{\epsilon_{\perp}} + \frac{k_z^2}{\epsilon_{\parallel}} = \frac{\tilde{S}^2}{c^2}$$

Since the length scale of the substructures is much smaller than the wavelength of light, one can define effective dielectric constants that control the macroscopic electromagnetic properties. The effective dielectric constants in the parallel and perpendicular directions for a one-dimensional metal-dielectric stack can be written as:

$\frac{1}{\epsilon_{\parallel}} = \frac{f_a}{\epsilon_a} + \frac{f_b}{\epsilon_b}$ and $\epsilon_{\perp} = f_a \epsilon_a + f_b \epsilon_b$, where f_a and f_b are the fill fractions and ϵ_a and ϵ_b are the dielectric constants of the two materials, respectively. By appropriate choice of fill fractions and the metallic and dielectric components, one has tremendous degree of control over the effective dielectric constants of the metamaterial structure. Closed iso-frequency surfaces different from a simple sphere (eg: ellipsoid) can occur in these metamaterials when $\epsilon_{\parallel} > 0$ and $\epsilon_{\perp} > 0$. On the other hand an extreme case of iso-frequency surface modification occurs when the dielectric constants show opposite sign ($\epsilon_{\parallel} < 0$ and $\epsilon_{\perp} > 0$) such that the iso-frequency curve opens up into a hyperbolic surface. This can be accomplished by controlling the fill fraction of the metal. The effective dielectric constants of one such metal dielectric structure as a function of wavelength is shown in **Fig.2**. The real part of the effective dielectric constant parallel to the layers (blue) goes through a sign change at 620 nm, while the perpendicular component (red) stays positive. This results in the hyperbolic dispersion. The shape of the dispersion curves are shown in the inset of **Fig. 2** for the different spectral regions.

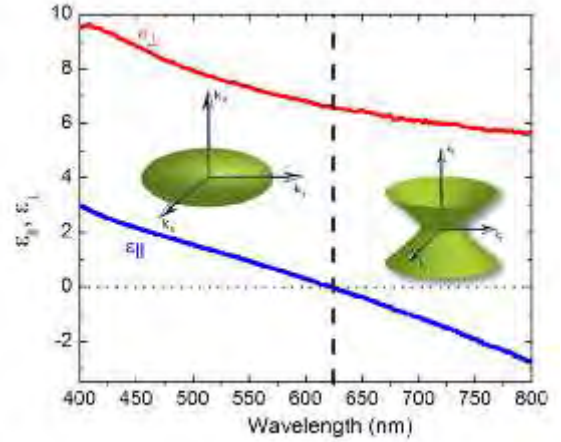


Fig. 2 Dispersion of a strongly anisotropic metamaterial exhibiting the elliptical and hyperbolic dispersion regimes.

One of the major issues in realizing HMMs with desired dispersion is the control over the layer thickness and the smoothness of the metal films. Unfortunately, at thickness less than 20 nm, silver films are usually percolated leading to island formation. This in turn affects the propagation length of surface plasmons at the metal-dielectric interface. Hence, one of the first issue we addressed under the program was to develop techniques to realize ultra-smooth silver films.

2. PROJECT DESCRIPTIONS

Growth of ultra-smooth silver films for layered hyperbolic metamaterials:

Growing thin (<20 nm) silver films on dielectric surfaces result in island formation since the thickness is below the percolation threshold. Scanning electron microscope image of a typical silver film, 10 nm in thickness grown on a glass substrate is shown in **Fig. 3a**. This film is highly percolated and hence mostly support localized plasmon modes. More recently, following and tweaking approaches reported previously [1, 2], we have been successful in demonstrating growth of ultra-smooth silver films on dielectric surfaces. The key here is to use a wetting layer which in our case was Ge. The structure consisted of Glass substrate/ Al_2O_3 /Ge/Ag. Scanning electron microscope image of the silver film grown using the Ge wetting layer is shown in **Fig. 3b** clearly indicating a smooth surface. **Fig. 3c** shows the atomic force microscope image of the surface of the. Less than 200 pm r.m.s roughness is observed in the Z (growth) direction and less than 2 nm r.m.s roughness in the x-y plane.



Fig. 3 Scanning electron microscope image of surface of silver film grown (a) without a wetting layer and (b) with a wetting layer of Ge. (c) Atomic Force Microscope image showing ultra-smooth surface topography of the film grown using the wetting layer.

Design and fabrication of bullseye grating structures for better in/out coupling:

One of the limiting aspects of HMM structures is the difficulty in coupling light into and out of the structures, thus making them difficult to implement in practical photonic device configurations. This issue arises due to the presence of high-k states in HMM structures which cannot couple to light in vacuum. One way to circumvent this issue while preserving the high-k states is to use a grating structure that couples light of specific k-vectors into and out of the structure. In this context we have recently designed a bullseye grating etched on to the metallic layer which efficiently out-couples the high-k states from the HMM into free space. Schematic drawing of the grating structure is shown in **Fig. 1**. Design of the grating structures were carried out using COMSOL finite element modeling package. Shown in **Fig. 4** is the simulated electromagnetic field profile along with the scattering when a radiating dipole is placed in the near field of the HMM. In the absence of the grating structure most of the light either gets absorbed or gets reflected (**Fig. 4a**). On the other hand when a grating is placed on the top layer, one can convert the in-plane surface

plasmon modes that are evanescent in nature into propagating modes (**Fig. 4b**). Shown in **Fig. 5 (a,b)** is the optimization of the grating period to out-couple a specific mode out of the HMM structure. Scanning electron microscope image of a bulls eye grating fabricated using focused ion beam etching is shown in **Fig. 5c**. The etching was carried out on structures where the silver film was not optimized for smoothness and hence one can see the rough surface on the grating. In the next phase of the project we are fabricating gratings on the optimized smooth silver films.

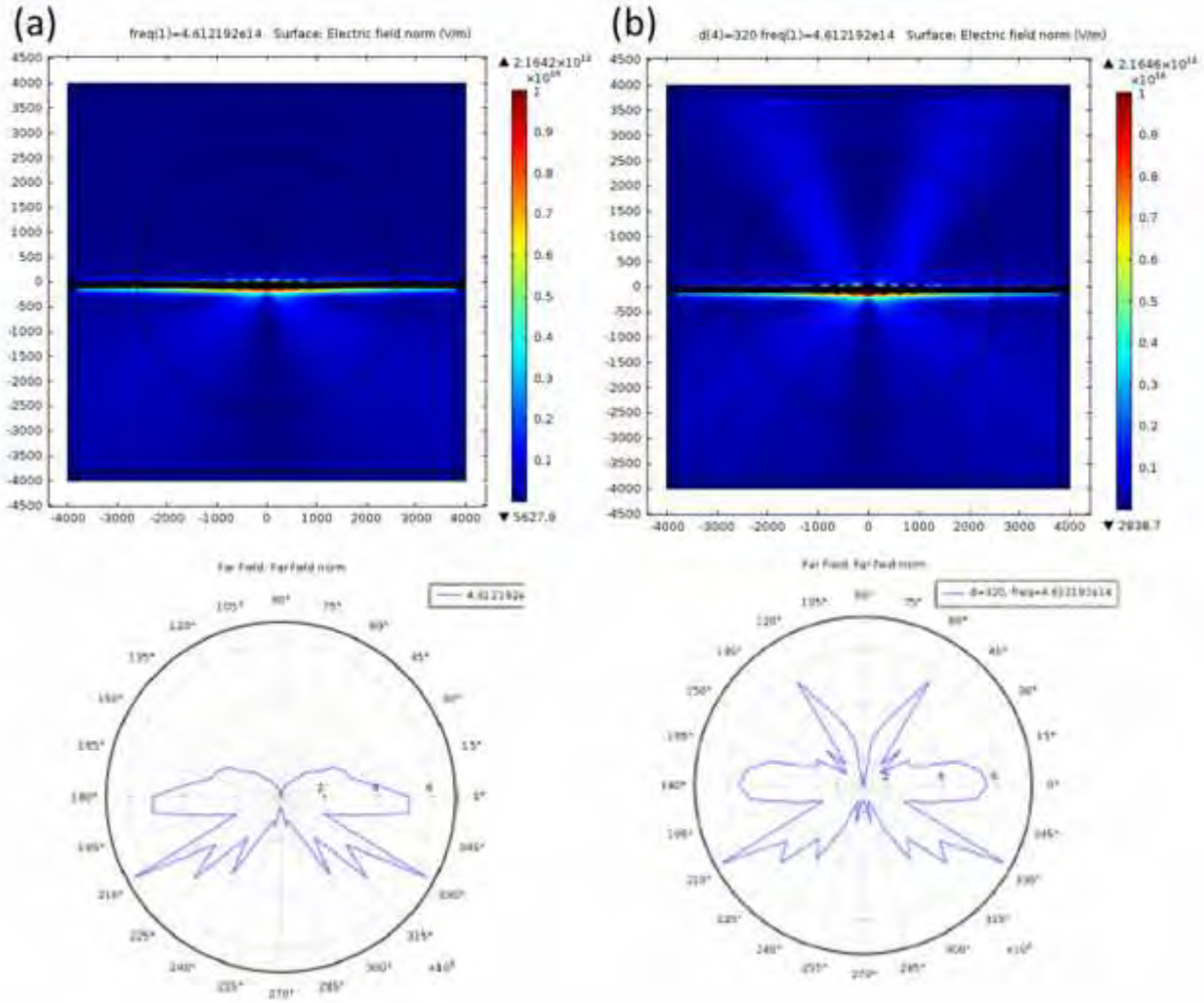


Fig. 4 Simulated electric field profile and scattering directions for a dipole placed in the near field of a HMM structure. (a) with no grating and (b) with a grating. It is clearly seen that in the presence of a grating, there is forward propagating waves that can be detected in the far field.

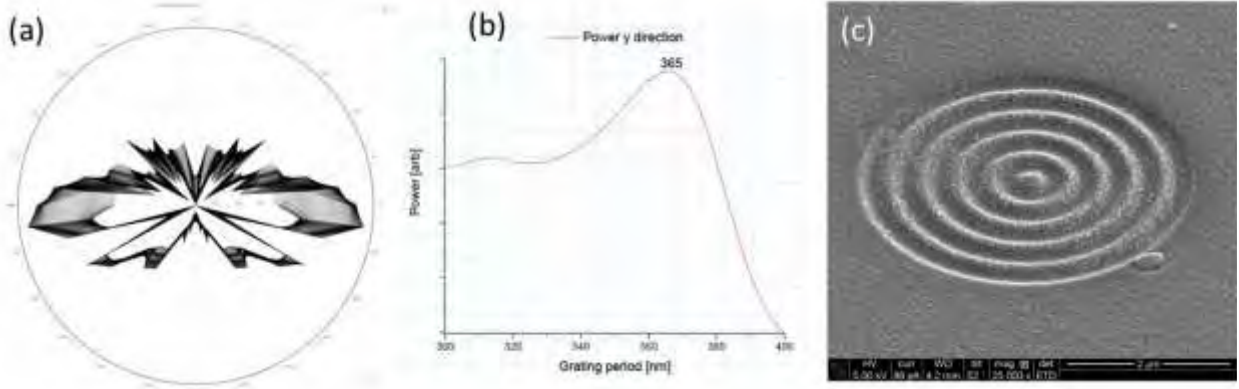


Fig. 5 Optimization of the grating period for maximum forward scattering amplitude. (a) polar plot showing the scattering angles and (b) the power at far field as a function of grating period. In this specific case for wavelength of 650 nm, we obtain maximum efficiency in out-coupling with a grating of 365 nm period. (c) SEM image of grating fabricated on a HMM structure using focused ion beam technique.

Fabrication of HMMs with ultrasmooth silver films and out-couplers

Using the results from the above two projects we fabricated the HMM structures with smooth silver films and germanium out-couplers. Shown in Fig. 6a is the schematic of the structure along with the cross-sectional TEM images of the layers showing smooth silver and alumina layers. The quantum dots are embedded inside the HMMs, Shown in Fig. 1c is the effective dielectric constants of the HMM structure realized. In the vicinity of the CdSe quantum dot emission, the metamaterial clearly has one component of its dielectric constant negative ($\epsilon_{||}$) and hence results in hyperbolic dispersion. Shown in Fig. 1d is the measured spontaneous emission lifetime of quantum dots on glass, a control sample of 1 unit cell, a 4 period structure and a 7 period structure where the quantum dots are embedded inside. Clearly the 7 period structure shows the most reduced lifetime as well as the narrowest distribution in lifetime. The reduced lifetime is due to enhancement in spontaneous emission rate. The reduced distribution in lifetime is

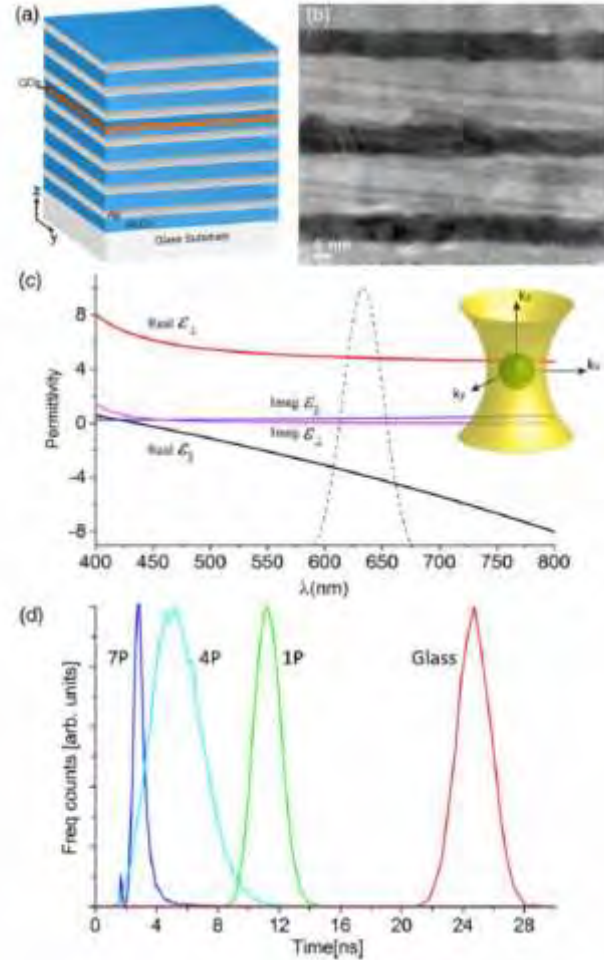


Fig. 6 (a) Schematic of the active HMM structure and (b) the cross sectional TEM image of the structure showing the alumina (light) and silver (dark) layers. (c) Effective dielectric constants of the active HMM structure. (d) Lifetime distributions of CdSe quantum dots embedded in different structures.

because the quantum dots being embedded inside the HMM allow almost identical coupling to the high-k modes of the HMM.

Finite element simulations using COMOSL were carried out to design the out-coupler. Shown in **Fig. 7** is the simulation showing the outcoupling from the optimized grating. Also for comparison the emission in the absence of a grating is also shown.

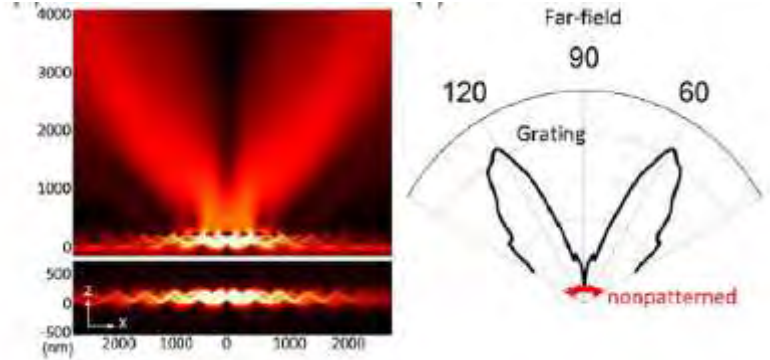


Fig. 7 (a) results of finite element simulations showing outcoupling using the bulls-eye grating structure.

Finally in **Fig. 8 (a)** we show the emission from active HMMs with different pitch gratings on top captured via confocal imaging. We see the 125 nm pitch to yield the most light output. The samples were pumped with a 440 nm laser and imaged using a scanning confocal imaging set

up with an avalanche photo diode (APD) detector. **Fig. 8(b)** shows the ratio of the emission intensity from the grating region to the background (unpatterned) of the active HMM. Clearly we see a factor of 20 enhancement in emission intensity for the 125 nm pitch grating. Shown in the inset of **Fig. 8(b)** is the lifetime map of the emission taken using a fluorescence lifetime imaging set

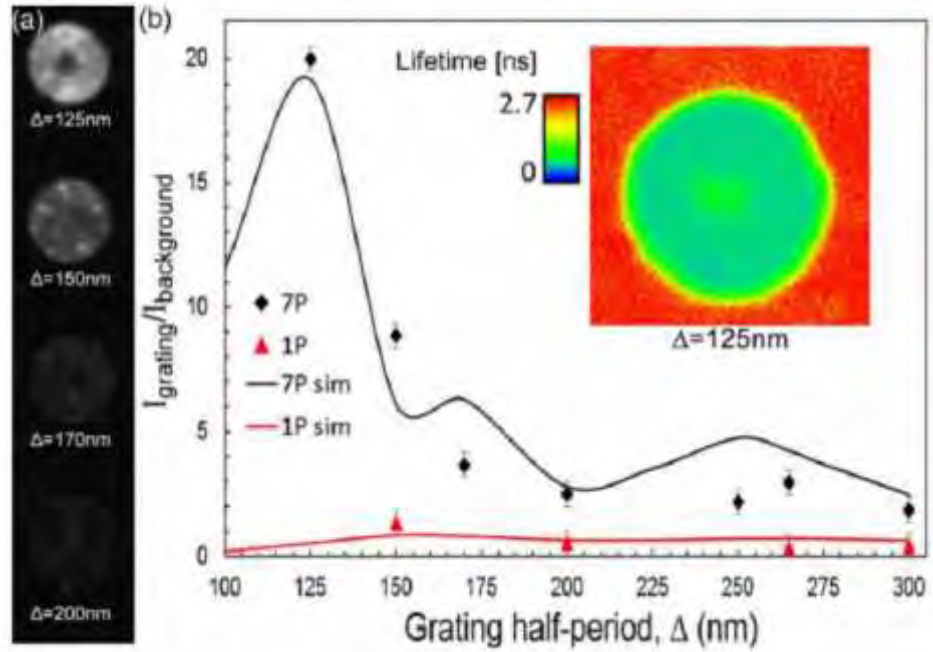


Fig. 8 (a) Scanning confocal image of the emission from the active HMM clearly showing enhanced emission from the 125 nm pitch grating structure. (b) Ratio of measured emission intensity at the grating to that of the background as a function of grating period. Inset: Fluorescence lifetime image of the structure showing decreased spontaneous emission lifetime from the grating region.

up. The lifetime of the emission is found to be shortest within the grating region. This is due to the more efficient coupling of vertical dipole emission which undergoes the greater Purcell enhancement to the grating. This work was published in *Optica* (2015) [3].

Design of sub-wavelength cavities using HMMs

In this part of the project we used finite difference time domain simulations to design sub-wavelength sized cavities made of HMMs that can efficiently confine light at high-k values. A typical structure in this case is shown in **Fig. 9a**. The structure consists of alternating layers of Ag and TiO₂. The total thickness of the structure is 30 nm and the lateral dimensions are 45 nm x 45 nm. This structure is shown to support a resonant mode at a wavelength of 2 μm (**Fig. 9b**) which is far larger than the physical dimensions of the structure. The electric field inside the HMM cavity along the different directions is shown in **Fig. 10**. It is clearly seen that the mode is well confined within the sub-wavelength HMM structure along all three dimensions.

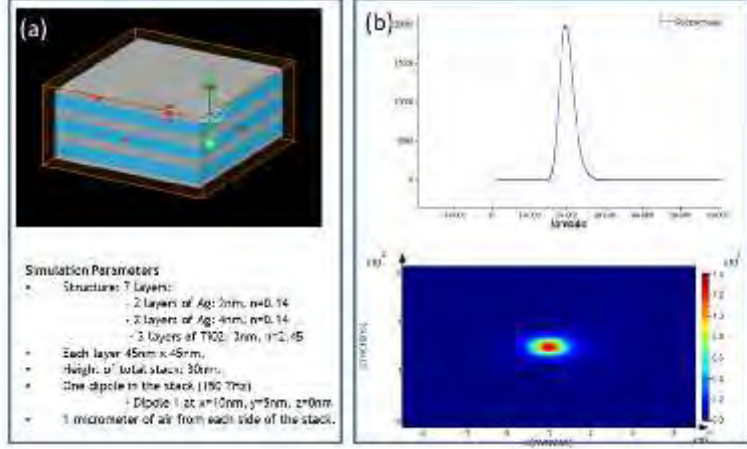


Fig. 9 (a) Schematic of the simulated sub-wavelength cavity structure along with simulation parameters and (b) simulation results showing resonant mode at 2 μm and the associated intensity profile inside the cavity at the resonance.

The electric field inside the HMM cavity along the different directions is shown in **Fig. 10**. It is clearly seen that the mode is well confined within the sub-wavelength HMM structure along all three dimensions.

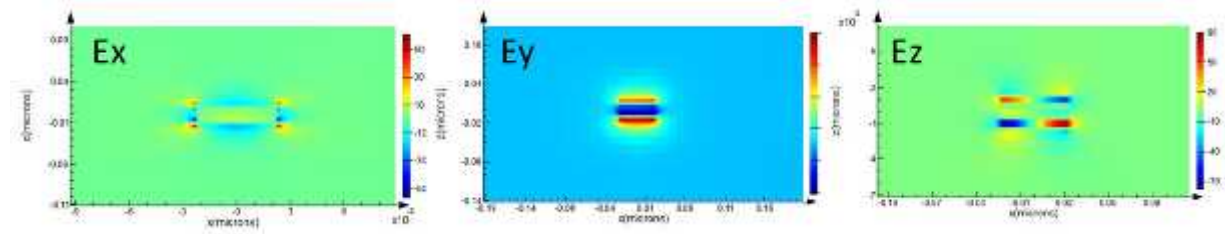


Fig. 10 Electric field simulations showing well confined modes in all three directions at 2 μm in the sub-wavelength structure.

3. REFERENCES

1. L. Vj, N. P. Kobayashi, M. S. Islam, W. Wu, P. Chaturvedi, N. X. Fang, S. Y. Wang, and R. S. Williams, "Ultrasooth Silver Thin Films Deposited with a Germanium Nucleation Layer," *Nano Lett.* **9**, 178 (2009).
2. W. Chen, M. D. Thoreson, S. Ishii, A. V. Kildishev, and V. M. Shalev, "Ultra-thin ultra-smooth low loss silver films on a germanium wetting layer," *Opt. Express* **18**, 5124 (2010).
3. "Active hyperbolic metamaterials: enhanced spontaneous emission and light extraction," T. Galfsky, H. N. S. Krishnamoorthy, W. Newman, E. E. Narimanov, Z. Jacob., and V. M. Menon,